Supporting Information

Atomistic Insights into Silicate Dissolution of Metakaolinite under Alkaline Conditions: Ab Initio Quantum Mechanical Investigation

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Models	ΔE_a (eV)	ΔE_a (kJ/mol)	E_reactant (eV)	E_transition state (eV)	E_product (eV)	ΔH (eV)	ΔH (kJ/mol)
(1)	1.13	109.03	-176.47	-175.34	-177.84	-1.37	-132.18
(2)	2.06	198.76	-178.18	-176.12	-177.46	0.72	69.47
(3)	1.18	113.85	-191.61	-190.43	-191.54	0.07	6.75
(4)	2.38	229.63	-189.71	-187.33	-191.54	-1.83	-176.56

Table S1. Results for four presented models with contribution of vdW interaction, hydrolysed by NaOH activator: energy barrier (activation energy) for hydrolysis reaction (ΔE_a = E_transition state – E_reactant), the energy change of reaction enthalpy (ΔH = E_product – E_reactant) obtained from first-principles based calculations.

Models	ΔE_a (eV)	ΔE_a (kJ/mol)	E_reactant (eV)	E_transition state (eV)	E_product (eV)	ΔH (eV)	ΔH (kJ/mol)
(1)	1.35	130.25	-175.88	-174.53	-176.94	-1.06	-102.27
(2)	2.08	200.69	-177.46	-175.38	-176.71	0.75	72.36
(3)	1.18	113.85	-190.65	-189.47	-190.70	-0.05	4.82
(4)	2.44	235.42	-188.81	-186.37	-190.70	-1.89	-182.36

Table S2. Results for four presented models without contribution of vdW interaction, hydrolysed by NaOH activator: energy barrier (activation energy) for hydrolysis reaction (ΔE_a = E_transition state – E_reactant), the energy change of reaction enthalpy (ΔH = E_product – E_reactant) obtained from first-principles based calculations.

Models	ΔE_a (eV)	ΔE_a (kJ/mol)	E_reactant (eV)	E_transition state (eV)	E_product (eV)	ΔH (eV)	ΔH (kJ/mol)
(1)	1.04	100.34	-176.63	-175.59	-178.17	-1.54	-148.58
(2)	1.76	169.81	-178.12	-176.36	-177.46	0.66	63.68
(3)	1.26	121.57	-192.20	-190.94	-191.25	0.95	91.66
(4)	2.15	207.44	-190.15	-188.00	-191.25	-1.10	-106.13

Table S3. Results for four presented models with contribution of vdW interaction, hydrolysed by KOH activator: energy barrier (activation energy) for hydrolysis reaction (ΔE_a = E_transition state – E_reactant), the energy change of reaction enthalpy (ΔH = E_product – E_reactant) obtained from first- principles based calculations.

Models	ΔE_a (eV)	ΔE_a (kJ/mol)	E_reactant (eV)	E_transition state (eV)	E_product (eV)	ΔH (eV)	ΔH (kJ/mol)
(1)	1.06	102.27	-175.83	-174.77	-177.32	-1.49	-143.76
(2)	1.80	173.67	-177.40	-175.60	-176.70	0.70	67.54
(3)	1.18	113.85	-191.11	-189.93	-190.24	0.87	83.94
(4)	2.2	212.27	-189.23	-187.03	-190.24	-1.01	-97.45

Table S4. Results for four presented models without contribution of vdW interaction, hydrolysed by KOH activator: energy barrier (activation energy) for hydrolysis reaction ($\Delta E_a = E$_transition state $- E$_reactant), the energy change of reaction enthalpy ($\Delta H = E$_product $- E$_reactant) obtained from first-principles based calculations.

Models	ΔE_a (eV)	ΔE_a (kJ/mol)	E_reactant (eV)	E_transition state (eV)	E_product (eV)	ΔH (eV)	ΔH (kJ/mol)
(1)	1.67	161.13	-178.87	-177.20	-178.68	0.19	18.33
(2)	0.88	84.90	-178.58	-177.70	-178.71	- 0.13	-12.54
(3)	1.62	156.30	-193.42	-191.80	-193.20	0.22	21.22
(4)	0.71	68.50	-193.62	-192.91	-193.20	0.42	40.52

Table S5. Results for four presented models with contribution of vdW interaction, hydrolysed by H_2O activator: energy barrier (activation energy) for hydrolysis reaction ($\Delta E_a = E$_transition state $- E$_reactant), the energy change of reaction enthalpy ($\Delta H = E$_product $- E$_reactant) obtained from first-principles based calculations.
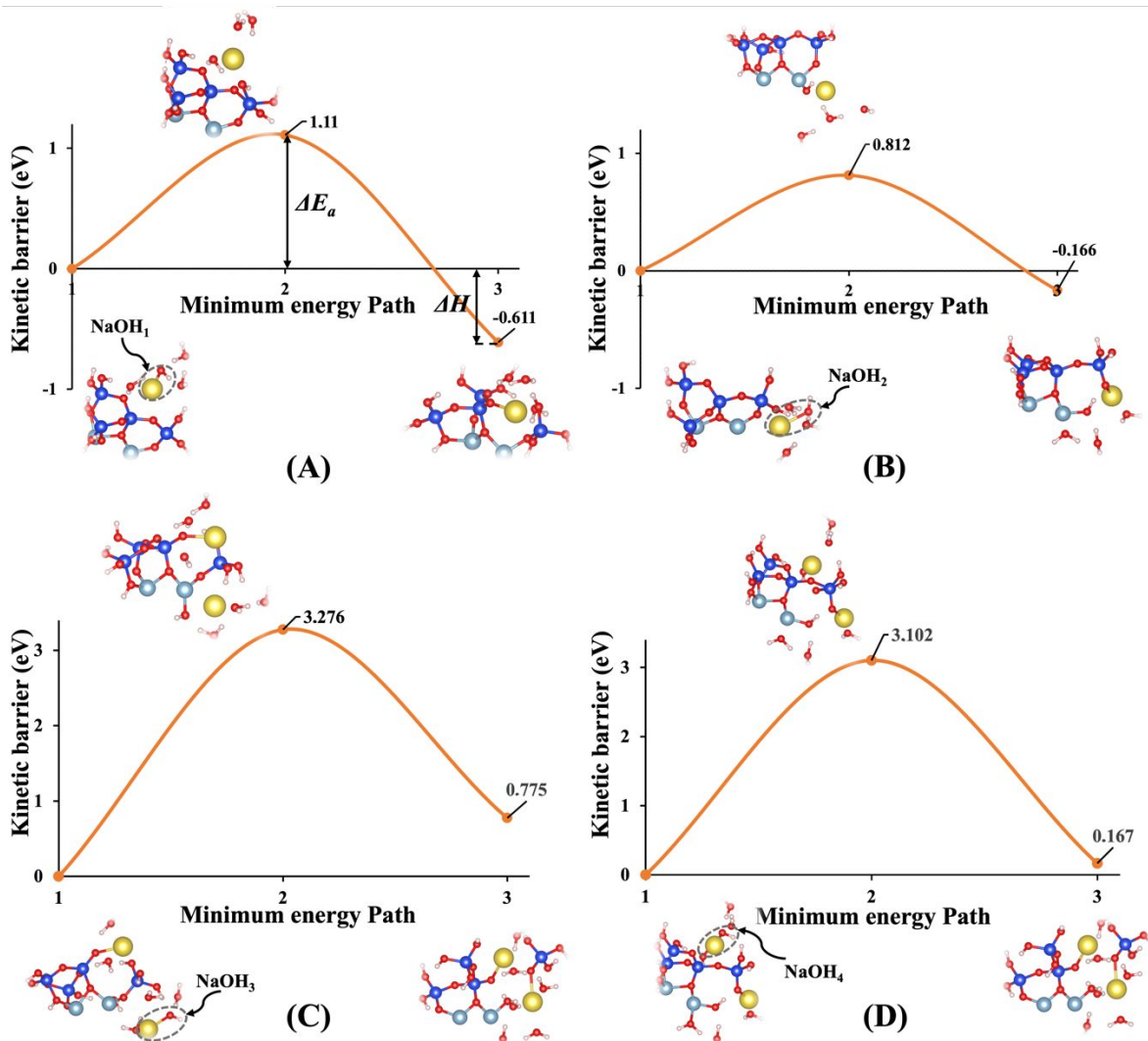
Figure S1. (A-D) Minimum energy path for computation of energy barrier (activation energy) of hydrolysis reaction (ΔEa), along with the associated energy change in reaction enthalpy (ΔH) including $(NaOH)_{1-4}$ absorbents surrounded by hydration shell without contribution of vdW interaction as presented for four models (1-4), respectively.
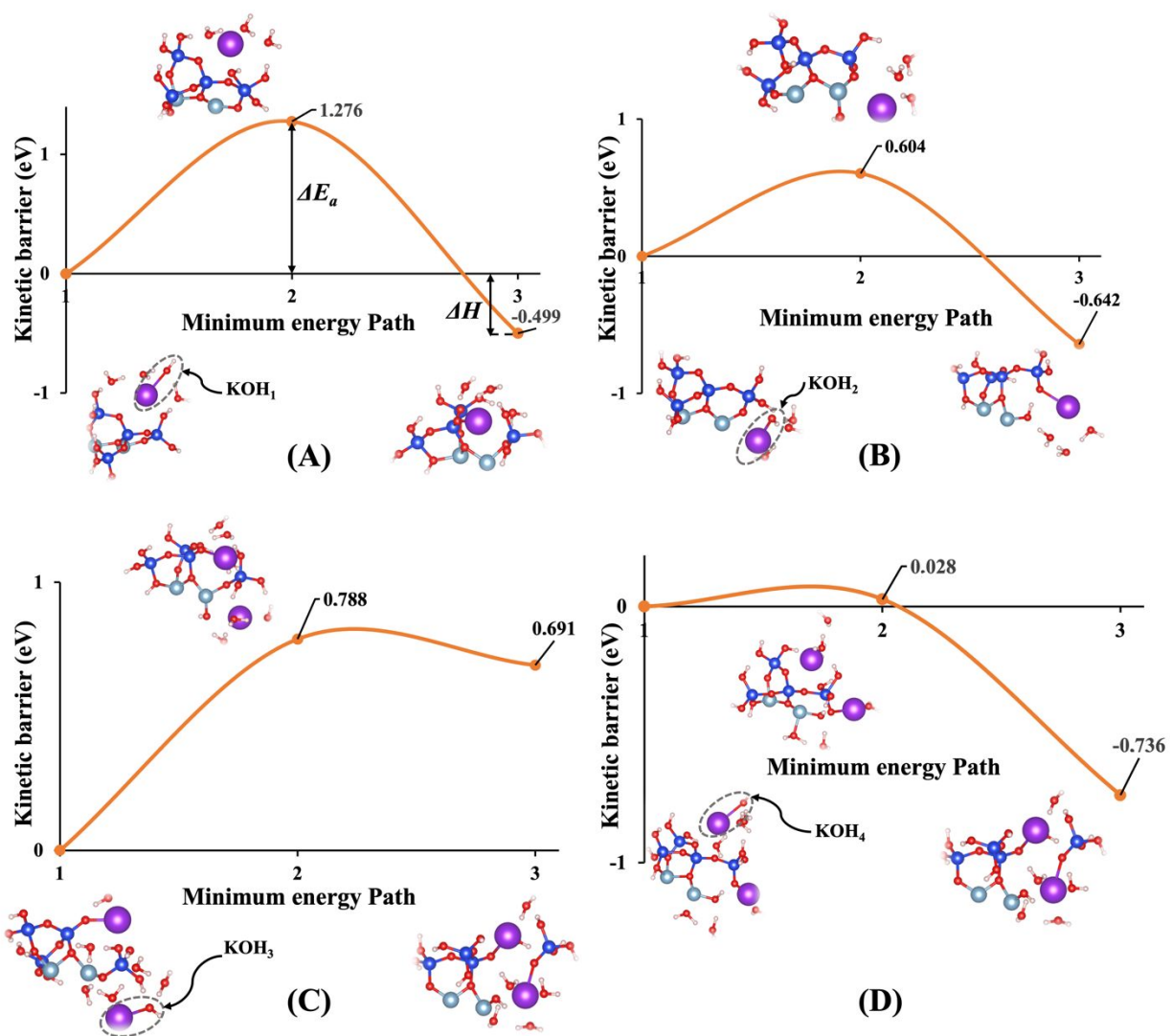
Figure S2. (A-D) Minimum energy path for computation of energy barrier (activation energy) of hydrolysis reaction (ΔEa), along with the associated energy change in reaction enthalpy (ΔH) including $(KOH)_{1-4}$ absorbents surrounded by hydration shell without contribution of vdW interaction as presented for four models (1-4), respectively.